Contaminant Mass Balance for Sinclair and Dyes Inlets, Puget Sound, WA

Eric Crecelius
> Battelle Marine Sciences Laboratory

Robert K. Johnston*, Jim Leather and **Joel Guerrero**
> Space and Naval Warfare Systems Center

Martin Miller and Jill Brandenberger
> Battelle Marine Sciences Laboratory

*Author to whom correspondence should be addressed: johnston@spawar.navy.mil

Abstract

Sinclair Inlet and Dyes inlets have historically received contaminates from military installations, industrial activities, municipal outfalls, and other nonpoint sources. For the purpose of determining a "total maximum daily load" (TMDL) of contaminants for the inlets, a contaminant mass balance for the sediments is being developed. Sediment cores and traps were collected from depositional areas of the inlets and surface sediment grabs were collected from fluvial deposits associated with major drainage areas into the Inlets. All sediment samples were screened using X-Ray fluorescence (XRF) for metals, and immunoassay for PAHs and PCBs. A subset of split-samples was analyzed using inductively coupled mass spectrometry (ICP/MS) for metals and gas chromatography mass spectrometry (GC/MS) for phthalates and PAHs, and GC electron capture detector (GC/ECD) for PCBs. Sediment cores were age-dated using radionuclides to determine the sedimentation rate and the history of sediment contamination. Streams and stormwater outfalls were sampled in both the wet and dry seasons to assess loading from the watershed. Seawater samples collected from the marine waters of the inlets and boundary passages to central Puget Sound were used to estimate the exchange of contaminates with central Puget Sound. The historical trends from the cores indicate that contamination was at a maximum in the mid 1900s and decreased significantly by the late 1990s. The thickness of the contaminated sediment is in the range of 15 to 45 cm. The initial drafting of the mass balance model for copper indicates that approximately 4,000 Kg/yr enters the inlets and 64% of this loading is from the leaching of boat and ship hulls. The removal of copper from the inlets is primarily a function of sedimentation (68%) and seawater advection into Central Puget Sound (32%).

Introduction

Sinclair and Dyes inlets, located in the Puget Sound west of Seattle, Washington (Figure 1), have historically received contaminants from military installations, industrial activities, municipal outfalls, and other non-point sources. Both inlets host Navy facilities, with the largest being the Puget Sound Naval Shipyard (PSNS) and Naval Station on Sinclair Inlet. A little over hundred years ago, Sinclair Inlet and the Kitsap Peninsula were relatively undeveloped. The Navy established a base in Sinclair Inlet in 1891, and in 1901 the Shipyard began operation and the town of Bremerton was founded (The Sun 2001). Rapid development in Bremerton and a boom in the population of Kitsap County followed major expansions at the Shipyard during World War I and World War II. At the height of World War II the population of Bremerton peaked at more than 80,000 people and industrial operations poured out goods for the war effort. Following the end of World War II, work at the Shipyard was reduced, but the Shipyard's workload remained high throughout the cold war and into the 1980s and1990s. In 1975, the Submarine Base at Bangor was established (Horn, Richard 1999), and since the late 1970s Kitsap County has experienced rapid growth in population, infrastructure, and development of open space. Currently, about a quarter of a million people live in Kitsap County (U.S. Census Bureau 2001). In 1998, the Washington State Department of Ecology listed some of the sediments in both inlets under Section 303(d) of the federal Clean Water Act because concentrations of mercury (Hg), copper (Cu), other metals, and organic chemicals exceeded sediment quality standards. To address environmental issues in the inlets and surrounding watershed, PSNS Project ENVironmental InVEStment (ENVVEST, PSNS Project ENVVEST 2002) was initiated as a cooperative effort among PSNS, regulatory agencies, and local stakeholders to evaluate sources, assess the mass balance, and develop "total maximum daily loads" (TMDLs) of contaminants for the inlets.
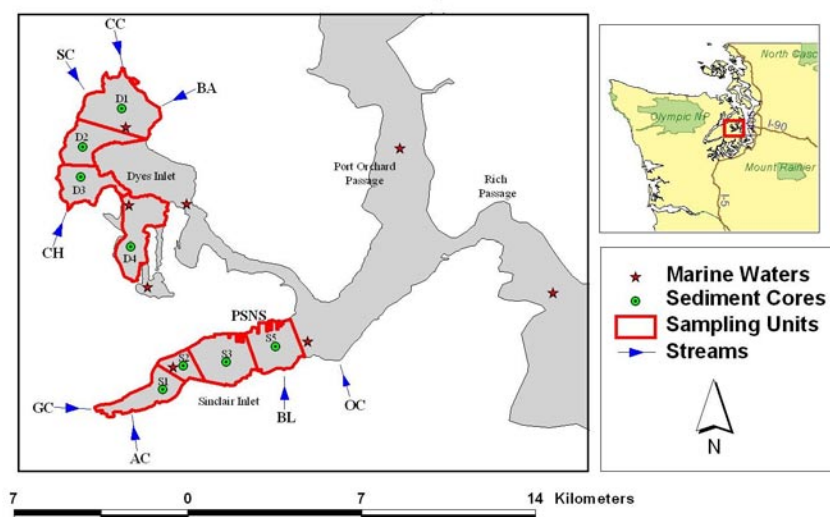
Figure 1. Sampling locations in Sinclair and Dyes inlets, Puget Sound, WA.

In this paper we present the methods and results used to characterize historical loading and estimate the current mass balance of contaminants in the inlets. The results obtained for Cu and Hg are used to illustrate the historical profile of contaminant loading in the estuary and the initial mass balance calculations for Cu are presented to demonstrate the technical approach. The complete set of findings and raw data will be published in a separate technical report (Crecelius et al. in prep).

Methods
In April 2002, sediment cores were collected from depositional zones within the inlets (Figure 1). These were age-dated to estimate the rate of removal of contaminants from the water column and to examine the historical trends in sediment contamination. Water and sediment samples were collected from streams entering the inlets and were analyzed to determine loadings from different land use areas. Seawater samples from the inlets were analyzed to estimate transport of contaminants between the inlets and into the Central Puget Sound. Because the inlets are hydrodynamically connected, contaminants discharged into one are shared with the other through tidal mixing; therefore, both inlets must be treated as one water body in terms of understanding environmental quality.

Sediment Coring
Sediment cores were collected at eight locations (Figure 1) using a piston corer supported in a frame and operated by SCUBA divers. This device recovered sediment cores with minimal disturbance or shortening compared with that caused by gravity coring. The core barrel had an inside diameter of 8.9 cm, a total length of 120 cm, and the length of recovered cores was about 100 cm each. The cores were kept upright during transportation and storage in a refrigerator. They were sectioned by extruding the sediment out the top of the core barrel using a piston inserted in the bottom of the core barrel. The top of the core (0-5 cm) was sectioned into two 2.5 cm sections and the remainder (5-100 cm) was sectioned into 5 cm sections.

Stream and Seawater Sampling
Water samples were collected during two seasons (wet – March 2002, and dry – September 2002) from eight streams draining into the inlets, and at eight marine water stations located within the inlets and passages, which exchanges with Central Puget Sound (Figure 1). Water samples were collected for trace metals analysis in pre-cleaned Teflon bottles using ultra-clean sampling techniques, following the U.S. Environmental Protection Agency (EPA) Method 1669 (EPA 1996a). All water samples were placed on ice in coolers and maintained at 4ºC while being transported to the laboratory. Half of each raw water sample was filtered through a 0.45-micron pre-cleaned filter in a Class 100 clean bench to produce the "dissolved" fraction.

Sediment Chemistry Screening

A split of approximately 50 gm of each sediment sample was screened using XRF for Cu, lead (Pb), iron (Fe), and zinc (Zn); and immunoassay for PAHs and PCBs (Kirtay and Apitz 2000). For metals analysis 10g of wet sediment were exposed to X-ray energy and the fluorescing spectrum was analyzed to quantify the metal ions present in the sample (U.S. EPA 1998). Approximately 10% of the samples analyzed using XRF were also analyzed using ICP-MS to validate the screening results.

Sediment Analyses

The sediment samples were homogenized and then split for metals and organics analysis. The metal sample split was freeze-dried and the ratio of wet-to-dry sediment was determined as the fraction of solids for each sample. Each freeze-dried sample was homogenized by milling, acid-digested, and then analyzed for Cu by ICP-MS and for Hg by cold vapor atomic absorption spectroscopy (CVAA). Sediment splits for organics were solvent-extracted and analyzed by GC/MS for phthalates, PAHs, and GC/ECD for PCBs (data not shown). Sediment digestion, extraction, and analysis methods for metals and organic compounds are provided in Lauenstein and Cantillo (1998).

Radionuclides Analysis

To determine sedimentation rates and the chronology of sediment contamination, subsamples of sediment core sections were analyzed for lead-210 (^{210}Pb) and cesium-137 (^{137}Cs). To prepare the sediment for alpha counting, about 3 g dry sediment was acid-digested, and polonium-210 (^{210}Po) was plated on a silver disk. The activity of ^{210}Pb was determined by counting the granddaughter ^{210}Po alpha particle on a silicon barrier diode detector, similar to the method of Koide et al. (1973). The activity of ^{137}Cs was determined by gamma-counting approximately 50 g dry sediment, using a germanium diode detector.

Sedimentation Rate

Sedimentation (cm/yr) and deposition (gm/cm^2/yr) rates were estimated using a steady-state ^{210}Pb dating technique (Lavelle et al.1986). This method assumes that (1) sedimentation rate is constant; (2) loss of ^{210}Po occurs only by radioactive decay; and (3) mixing is confined to the surface mixed layer. The year of deposition for the midpoint of each core section was calculated. The relationship between sediment wet density and sediment dry weight was used to calculate the deposition rate. The location of the subsurface ^{137}Cs peak in each core was used to evaluate the accuracy of the ^{210}Pb core dating technique.

Water Analyses

Ultra-clean trace metal sampling and analysis techniques were followed for metals in water samples (U.S. EPA 1996a, b, c). All water samples were analyzed for Hg using cold vapor atomic fluorescence spectrometry (CVAF) in accordance with EPA Method 1631, Revision E (EPA 2002). Freshwater samples for all other metals (e.g., Cu for this paper) were analyzed using ICP-MS, in accordance with EPA Method 1638 (EPA 1996b). Seawater samples were preconcentrated using Fe and palladium (Pd) prereduction, in accordance with the Battelle Marine Sciences Laboratory (BMSL) standard operating procedure (SOP) for seawater preconcentration, which is derived from modifications to EPA Method 1640 (EPA 1996c). The preconcentrated samples were then analyzed by ICP-MS.

Results

Sediment Screening Results

The analytical results obtained for Cu, Pb, and Zn showed good agreement between XRF and ICP-MS (Figure 2). The rapid estimates of contaminant concentrations from the screening analysis allowed the more expensive, low-detection-level chemistry analysis by ICP-MS to be focused on selected samples. This was particularly useful for the core subsections, because preindustrial background values for Pb were identified in the screening methods, and subsections deeper than those showing stabilization of Pb at background levels were not submitted for ICP-MS analysis.

Temporal Trends from Dated Cores

The historical trends from the cores indicate an increase in the contaminant levels of Hg, Cu, PAHs, and PCB beginning around 1900. The significant increase in these contaminants peaked between 1940 and 1960. The results obtained for Cu and Hg are used to illustrate the historical profile determined from the data (Figures 3 and 4). The thickness of the layer of contaminated sediment ranged from 15 cm to 50 cm. These trends, or peaks and declines, of contaminants
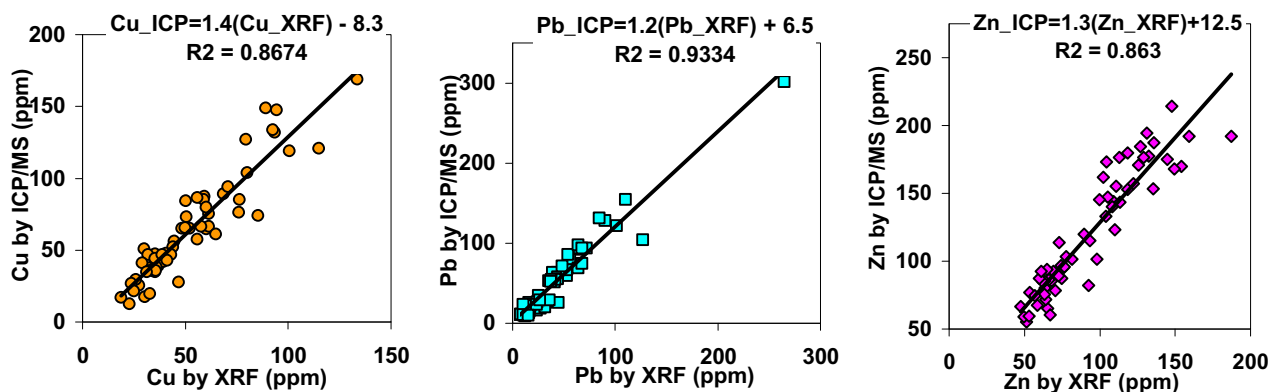
Figure 2. The relationship between Cu, Pb, and Zn measured by XRF and ICP-MS obtained for sediment samples.

in the Sinclair and Dyes Inlet cores accurately reflect the known history of both the uses of these contaminants and environmental regulations. In the 1970s, Congress created the Environment Protection Agency (EPA) and enacted environmental regulations to control pollution sources. The enforcement of environmental laws is reflected in the core profiles, which shows a significant decline in the later part of the 20th Century. The average sedimentation rate for the eight core profiles was estimated to be 0.115 ± 0.051 g/cm²/y; with the average thickness of the annually deposited sediment estimated to be 0.25 ± 0.12 cm/yr for the inlets.

Historical uses of Hg date far earlier than the 1930s; therefore, the initial increase of Hg is more diffuse in the cores than the profiles for the other chemicals. The peak in Hg for both Sinclair and Dyes inlets occurred in the 1940s (Figure 3), corresponding to the escalation of industrial activities associated with World War II. However, the reduction of Hg used by the Navy and the passage of regulations designed to minimize Hg discharges are also reflected in the core profiles, in which Hg levels decrease dramatically after the peak in the 1940s. Present-day sediment quality is improving; however, Hg is still slightly above the Washington State sediment quality standard (SQS) of 0.41 µg/g.

The increase in Cu concentrations in the sediments of the inlets corresponded to the onset of military operations in the early 1900s (Figure 4). The preindustrial concentration of Cu in sediment from the inlets was 30 to 40 µg/g. The maximum input of Cu to the inlets occurred in the 1960s, and there has been a significant decrease since then (Figure 4). Copper has been and continues to be used in antifouling paint on Navy, private, and commercial vessels. However, the practices for maintaining vessels have changed in the last few decades to minimize the release of Cu during hull cleaning and painting.

Copper Mass Balance
We calculated a mass balance for Cu in the inlets to compare the rates of input from different sources with the rates of removal. Only a mass balance for Cu is presented in this paper because there are very limited data for other contaminants at this time. Even the data available for the Cu mass balance are limited. However, because there are reasonable estimates for many sources of Cu to the inlets (Johnson and Grovhoug 1999), we believe that the estimated mass balance will be instructive in refining the mass balance for Cu and other contaminants. We have assumed that the only removal processes for Cu are burial in the fine-grain sediments of the inlets and advection of seawater from the inlets into Central Puget Sound.
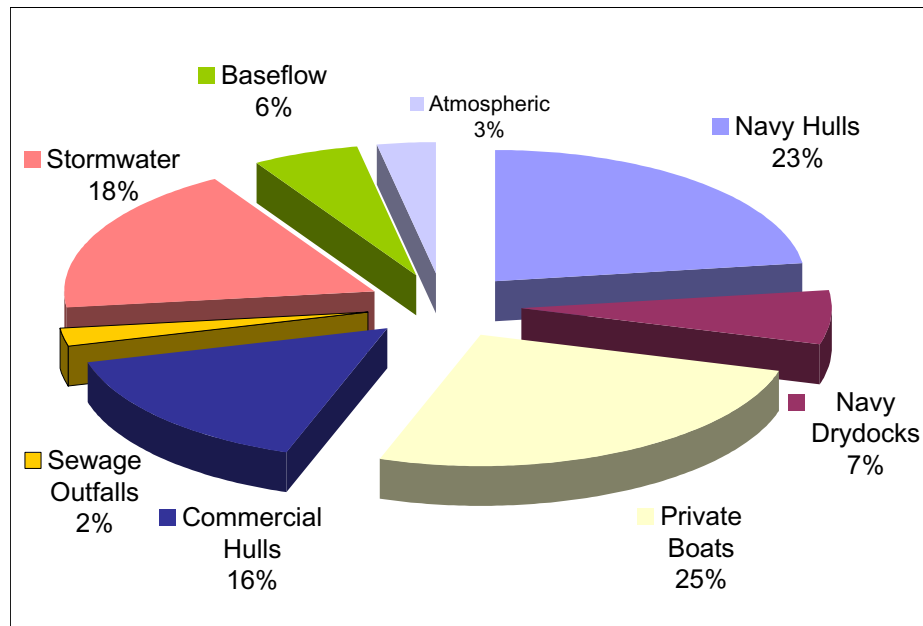
Sources of Copper
Sources of Cu include Navy hulls, Navy dry-docks, private boat hulls, commercial hulls, discharge from storm events, base flow of streams, atmospheric deposition, and sewage outfalls. Most of the estimates of Cu input were obtained from Johnson and Grovhoug (1999) and P.F. Seligman (Space and Naval Warfare Systems Center – SSC, San Diego, CA, personal communication, 2003). The annual Cu inputs were: Navy hull leaching 980 kg/yr; Navy dry-dock discharge water 291 kg/yr; private boat hull leaching 1104 kg/yr; commercial vessel hull leaching 670 kg/yr; storm-event runoff 792 kg/yr; base flow 238 kg/yr; atmospheric deposition 132 kg/yr; and sewage outfalls 98 kg/yr (Figure 5). Hull leaching from both Navy and private vessels accounted for 48% of the Cu sources to the inlets. The sum of the individual sources provided an annual loading of Cu to the inlets of 4,305 kg/yr.
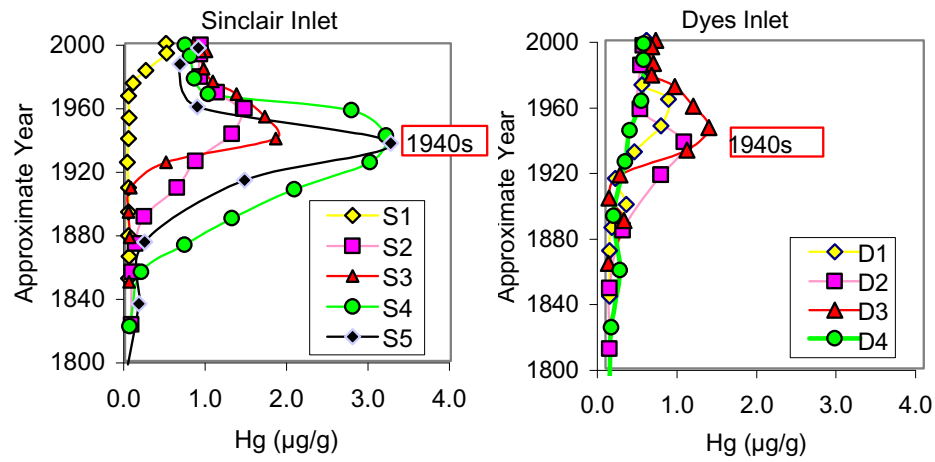
Figure 3. Core profiles for Hg in Sinclair and Dyes inlets.
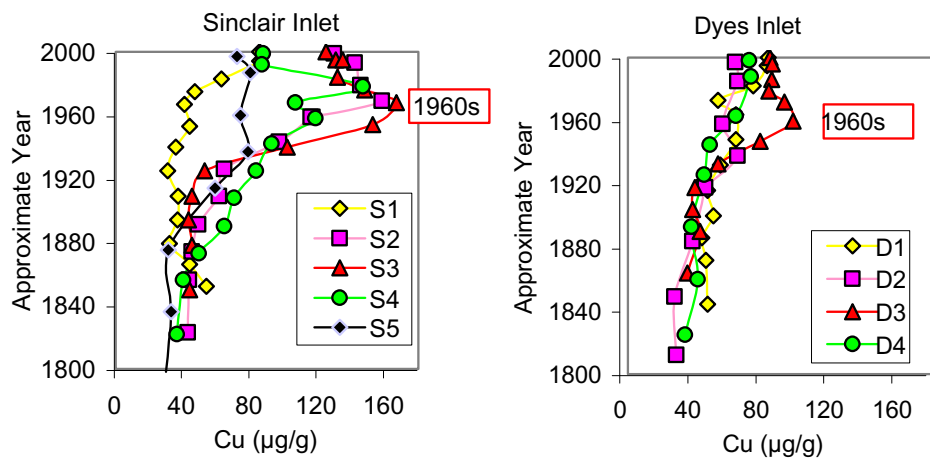
Figure 4. Core profiles for Cu in Sinclair and Dyes inlets.

Figure 5. Copper sources in Sinclair and Dyes inlets.

We estimated Cu loading from the base flow of streams and atmospheric deposition. The input of copper to the inlets was estimated for eight streams during base-flow conditions representing non-storm events. Stream samples were collected during the wet (March 2002) and dry (September 2002) seasons. Table 1 provides the six-month mean stream flows and Cu concentrations used to calculate a Cu loading for each stream during baseflow wet and dry seasons. The total Cu concentrations range from 0.3 to 15.7 µg/L. The available stream-flow data were used to calculate the Cu loading for each stream during wet and dry base-flow conditions: 225 kg/yr copper and 13 kg/yr, respectively for a total of 238 kg/yr.

**Table 1.** Baseflow Loading for Primary Streams Entering Sinclair/Dyes inlets.

Stream Name	Code	Baseflow Wet Season			Baseflow Dry Season		
		6 Month Mean Flow (m³/s)	Cu Concentration (µg/L)	Cu Loading (Kg)	6 Month Mean Flow (m³/s)	Cu Concentration (µg/L)	Cu Loading (Kg)
Clear Creek	CC	0.539	2.0	17	0.152	0.33	0.79
Strawberry Creek	SC	0.377	1.9	12	0.0644	0.47	0.48
Barker Creek	BA	0.264	2.5	10	0.0994	0.31	0.50
Chico Creek	CH	1.13	2.5	44	0.224	0.60	2.2
Gorst Creek	GC	1.27	1.5	29	0.355	1.1	6.3
Blackjack Creek	BL	0.476	1.5	12	0.259	0.26	1.1
Olney Creek	OC	0.197	9.9	31	0.108	0.47	0.81
Anderson Creek	AC	0.283	16	70	0.107	0.26	0.45
		Total Wet Season Load		225	Total Dry Season Load		13

Measurements have not been made of the atmospheric deposition near the inlets. Crecelius (1981, 1991) measured the deposition of Cu and other contaminants at several locations in Western Washington, including five sites around Commencement Bay in Tacoma, near Sequim Bay, and at Quillayute near the northwestern Washington coast. The Cu deposition rate at these sites ranged from about 1 to 54 mg/m^2/yr. The deposition rate for Riverside School in the Puyallup River valley, WA was 20 mg/m^2/yr, which could be a reasonable estimate for the inlets as well. Applying this rate to the area of sediment deposition (18.1 km^2) in the inlets results in an annual flux of 132 kg. The estimate from Johnson and Grovhoug (1999) was considerable lower (3 kg/yr) and was based on a deposition rate from the tropical North Pacific. Further information about atmospheric deposition may be obtained from the analysis of sediment cores obtained from Heinz Lake, located within the Sinclair Inlet watershed but isolated from all sources of contamination, except atmospheric deposition (Crecelius et al. in prep).

The estimate for leaching of dissolved Cu (1104 kg/yr) from private boats in the inlets was provided by P.F. Seligman (SSC, personal communication, 2003). He estimated that there were 1148 boats moored year-around in the marinas located in Sinclair and Dyes inlets. The Cu-leaching rate was estimated from measurement on pleasure boats in San Diego (Valkirs et al. 2003-in press). In comparison, the release rate of total Cu from two Puget Sound marinas was reported by Crecelius (1989) to be 33.9 kg/yr for a marina with 450 boats and 146 kg/yr for a marina containing 950 boats. If these release rates are adjusted to 1148 boats, the annual input to the inlets would be either 87 kg/yr or 176 kg/yr, either of which is much lower than Seligman's estimate, and could have to do with the differences in types of paint, the water temperature, and boat usage between Puget Sound and San Diego Bay.

Seawater Export
The transport of Cu out of the inlets by advection is based on the difference in the concentrations of total Cu in the inlets and at the boundary of the inlets, and the volume of seawater exchange between the inlets and Central Puget Sound. Since the concentration of Cu is higher in the water inside than that outside the inlets, the net transport of Cu will be out of the inlets into Central Puget Sound. The rate of exchange of seawater in the inlets was provided by P.F. Wang (SSC, San Diego, CA, personal communication, 2003). In the process of completing a hydrodynamic transport model for the inlets, he estimated that the flushing time for Sinclair Inlet was 12 days with a basin volume of 100.9 million m^3, and for Dyes Inlet 15 days with a basin volume of 155.6 million m^3. These flushing rates are 3.07 billion m^3/yr for Sinclair and 3.79 billion m^3/yr for Dyes Inlet, or a combined transport of 6.86 billion m^3/yr.
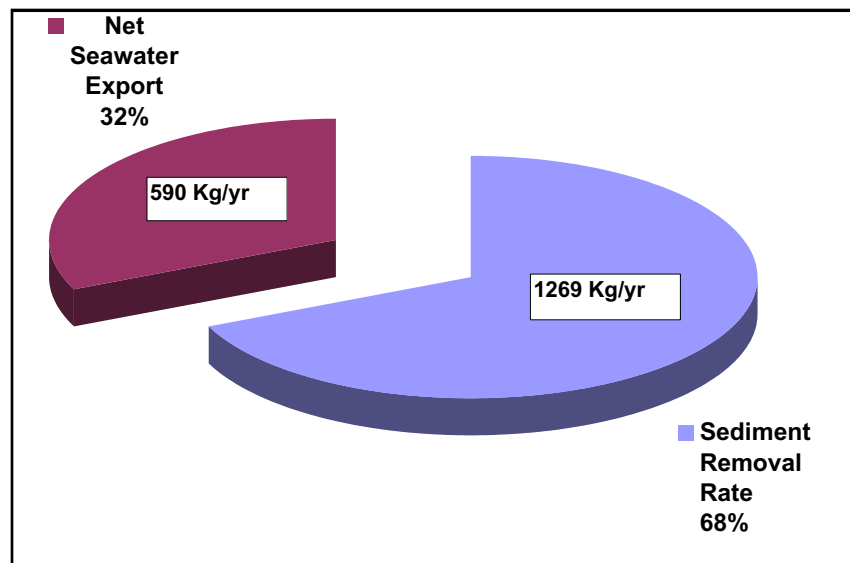
Figure 6. Fate of Cu in Sinclair and Dyes inlets.

The concentration of total Cu in the surface water in the inlets was determined to be 0.692 µg/L in March 2002 (wet season base flow) and 0.703 µg/L in September 2002 (dry season base flow). The total concentration of Cu in seawater at the margins of the hydrodynamic model—that is, at the model boundary sampling stations in Port Orchard and Rich Passages —was 0.581 µg/L in the wet season and 0.643 µg/L in the dry season. The mean annual total Cu concentration in the inlets was 0.695 µg/L, and the mean Cu concentration at the boundary is 0.612 µg/L, resulting in a difference of 0.083 µg/L. The annual seawater exchange between the boundary and the inlets is 6.86 billion m^3, based on the flushing rates of the two inlets. This results in the net annual transport of 590 kg Cu from the inlets into Central Puget Sound.

Copper Loading to Sediment
The removal of contaminants from the water column to the sediments was estimated from the sedimentation rate and the concentrations of contaminants in the surface sediment (0-2.5 cm) and is illustrated in Figure 6. We assumed that the piston core collections in April 2002 were representative of the depositional rates for specific areas of the inlets, and that the concentration of contaminants in the surface of these cores was representative of the specific areas. Each Inlet was divided into four sediment units, and each unit (Figure 1) contained one core that was used to estimate the sediment accumulation rate. The area of the unit was based on the 20-foot MLLW contour and sediment from water depths shallower than this contour was assumed to have very low sediment accumulation rates. The sediment accumulation rates ranged from 0.06 $g/cm^2/yr$ to 0.20 $g/cm^2/yr$ dry sediment. The concentrations of Cu in the surface sediment ranged from 63 µg/g to 130 µg/g dry weight. The area of the units ranged from 0.7 to 2.5 km^2. The Cu loading to the sediment ranged from 36 kg to 443 kg; providing a total sediment removal rate of 747 kg/yr for Sinclair Inlet and 522 kg/yr for Dyes Inlet. Since the inlets were treated as one unit, the sum of the removal rates for each inlet was added to the mass balance model to provide the total basin sediment removal rate for Cu of 1269 kg/yr or 68 % of the Cu removal in the inlets.

Discussion
The similarity between the historic trends, identified in the core profiles, for Sinclair and Dyes inlets substantiates the hypothesis that the inlets should be treated as one system. In addition, the presence of similar contaminants at a lower concentration in Dyes Inlet sediment relative to Sinclair Inlet, suggests that Sinclair Inlet is the major source for the contaminants reported in this study. The Cu mass balance is considered a work in progress; therefore additional studies are currently in progress or under consideration. The major contribution (~50%) of Cu to the total sources in the inlets is from Cu-based antifouling paint on boats (commercial, private, and Navy). Additional verification of these loadings could significantly improve the mass balance. For example, we recommend site-specific hull Cu release rates be determined in Puget Sound or Sinclair Inlet because of the large discrepancy between field measurements in Puget Sound and estimates made for San Diego Bay. The Cu budget developed for San Diego Bay also showed that about 50% of the Cu loading came from civilian (38%) and Navy (11%) hull leaching (Chadwick et al. in review). The total loading of Cu in San Diego Bay (17,000 kg/yr) was about 4 times higher than the Cu loading calculated for Sinclair and Dyes inlets

with 42% being retained in the sediments and 58% exported to the Pacific Ocean (Chadwick et al. in review). Owing to the high loading, San Diego Bay has much higher Cu concentrations in the water column, exceeding water quality criteria (3.1 µg/L) in significant areas of the central bay and within enclosed yacht basins (6.0 µg/L, Chadwick et al. in review).

Based on our calculations, two thirds of the Cu entering the inlets is deposited in the sediment (Figure 6). The Cu removal rates for seawater advection and sediment deposition were estimated from only a few chemical analyses and these fate mechanisms could be greatly improved by additional confirmatory sampling. Our analysis suggests that the sediments of Sinclair and Dyes inlets retain more of the total Cu load than do the sediments of San Diego Bay. If the sedimentation rates and Cu concentrations in sediment are lower near the edge of the sediment units compared to the locations of the cores, then deposition rates may be too high. Currently, loading from streams during storm events is being evaluated and surface sediment sampling in the inlets and storm event sampling for storm water outfalls is planned. Additional fieldwork is needed to determine the concentration of contaminants in the ebb tide from the inlets and the flushing rates in order to refine the fate term in the mass balance. Investigations on the contaminant levels in ecologically abundant marine organisms should also be conducted to determine the fraction of chemicals that are biologically active and estimate the mass of chemical contained within the inlets' biota.

Conclusions

A study of the mass balance of contaminants in Sinclair and Dyes inlets is being conducted to support development of TMDLs and increase the understanding of how the ecological system functions. Estimates of sedimentation rates and historical tends of contaminant loading in the estuary were obtained from analysis of sediment cores taken in depositional area of the inlets. Concentrations of contaminants in streams and marine waters of the inlets were used to estimate base flow loading and net exchange with the central part of Puget Sound. Sedimentation rates ranged from 0.06 g/cm^2/yr to 0.20 g/cm^2/yr (dry sediment) and the historical trends from the cores indicated that contamination was at a maximum in the mid 1900s and decreased significantly by the late 1990s. The thickness of the layer of contaminated sediment ranged from 15 cm to 45 cm. The initial mass balance calculations for Cu showed that about half of the copper loading was from antifouling paints on civilian and Navy ships, about 68% of the total Cu load is retained in the sediments, and 32% of the load is exported to the Central Puget Sound. The major sources of uncertainty were the lack of data on hull leach rates of Cu in the Puget Sound, loading from storm events, and atmospheric deposition and the scarcity of data used to characterize tidal exchange and sediment contamination levels.

Acknowledgements

Bibliography

Chadwick, DB, I Rivera-Duarte. P.F. Wang, and A. Zirino, (2003-in review). Modeling the Mass balance and Fate of Copper in San Diego Bay, California, *Limnology and Oceanography.*

Crecelius, E. A., 1981, Prediction of Marine Atmospheric Deposition Rates Using Total [7]Be Deposition Velocities, *Atmospheric Environment*, **15**: 579-582.

Crecelius, E. A., T. J. Fortman, S. L. Kiesser, C. W. Apts and O. A. Cotter, 1989, Survey of Contaminants in Two Puget Sound Marinas, USEPA Region 10 Report 910-9-89-014, Seattle, Washington.

Crecelius, E. A., 1991, Estimate of the Atmospheric Deposition of Contaminants on Commencement Bay, Washington, 84th Annual Meeting and Exhibition Proceedings, Vancouver, British Columbia.

Crecelius, E., et al. (in prep). Contaminant Mass Balance for Sinclair and Dyes inlets, Puget Sound, WA. PSNS Project ENVVEST, Final Report.

Horn, Richard 1999. The Bangor Boom. By Published in The Sun: 12/30/1999. http://www.thesunlink.com/news/99december/daily/1230a1b.html

Johnson, H. D. and J. G. Grovhoug, 1999, *Copper Loading to U. S. Navy Harbors Bremerton, WA*, SPAWAR Systems Center Technical Document 3052, San Diego.

Kirtay, V. J. and S. E. Apitz, 2000. Rapid Sediment Characterization (RSC). Navy Guidance for Conducting Ecological Risk Assessments: Issue Papers. Space and Naval Warfare Systems Center, San Diego, CA. 22pp. http://web.ead.anl.gov/ecorisk/issue/pdf/rsc.pdf

Koide, M. K., E. Bruland and E. D. Goldberg, 1973, Th-228/Th-232 and Pb-210 Geochronologies in Marine and Lake Sediments, *Geochemica et Cosmochimica Acta*, **37**: 1171-1183.

Lauenstein, G. G. and A. Y. Cantillo (eds.), 1998, Sampling and Analytical Methods of the National Status and Trends Program Mussel Watch Project: 1993-1996 Update, NOAA Technical Memorandum NOS ORCA 130, Silver Spring, MD.

Lavelle, J. W., G. J. Massoth and E. A. Crecelius, 1986, Accumulation Rates of Recent Sediments in Puget Sound, Washington, *Marine Geology*, **72**, 59-70.

PSNS ENVVEST 2002. PSNS Project ENVVES: Technical Work Master Plan (REVIEW DRAFT), Puget Sound Naval Shipyard, US Environmental Protection Agency, and Washington State Department of Ecology, May 10, 2002.

The Sun 2001. Bremerton Centennial 1901-2001: A historical retrospective. http://www.thesunlink.com/bremertoncentennial

U. S. Environmental Protection Agency, 1996a. Method 1669: Sampling Ambient Water for Determination of Metals at EPA Water Quality Criteria Levels, Office of Water Engineering and Analysis Division, Washington, D.C.

U. S. Environmental Protection Agency, 1996b. Method 1638: Determination of Trace Elements in Ambient Waters by Inductively Coupled Plasma - Mass Spectrometry, Office of Water Engineering and Analysis Division, Washington, D.C.

U. S. Environmental Protection Agency, 1996c. Method 1640: Determination of Trace Elements in Ambient Waters by On-Line Chelation Preconcentration and Inductively Coupled Plasma-Mass Spectrometry, Office of Water Engineering and Analysis Division, Washington, D.C.

U.S. EPA 1998. Method 6200: Field portable x-ray fluorescence spectrometry for the determination of elemental concentrations in soil and sediment. Office of Solid Waste and Emergency Response. Rev. 0 http://www.epa.gov/epaoswer/hazwaste/test/pdfs/6200.pdf

U. S. Environmental Protection Agency, 2002. Method 1631, Revision E: Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry, Office of Water Engineering and Analysis Division, Washington, D.C.

U.S. Census Bureau 2001. U.S. Census 2000. http://www.census.gov/dmd/www/2khome.htm

Valkirs, A.O., P.F. Seligman, E. Haslbeck, and J.S. Caso, (2003- in press), Measurement of copper release rates from antifouling paint under laboratory and *in-situ* conditions: implications for loading estimation of marine water bodies, *Marine Pollution Bulletin*.